THREE DIMENSIONAL INTEGRATED CHARACTERIZATION
AND ARCHIVING SYSTEM (3D-ICAS)

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ABSTRACT

The objective of this project is to develop a remote system that performs rapid in situ analysis of hazardous organic and radionuclide contamination on structural materials. Characterization sampling and analysis for hazardous organic and radionuclide contamination on concrete, asbestos, and transite for decontamination and decommissioning (D&D) is time-consuming, expensive, and has the potential to of expose humans to radiation and hazardous materials. Exposure limits, rest breaks, and donning and removal of protective garments generate waste in the form of contaminated protective garments and equipment. Survey times are increased and handling and transporting potentially hazardous materials incur additional costs. Off-site laboratory analysis is expensive and time-consuming, often necessitating delay of further activities until results are received.

The Three Dimensional Integrated Characterization and Archiving System (3D-ICAS) was developed to alleviate some of these problems. 3D-ICAS provides a flexible system for physical, chemical and nuclear measurements reduces costs and improves data quality. Operationally, 3D-ICAS performs real-time determinations of hazardous and toxic contamination. A prototype demonstration unit is available for use in early 2000.

I. INTRODUCTION

Accurate physical characterization of surfaces and the radioactive and organic contamination is a critical D&D task. Before clean-up operations can begin the site must be characterized with respect to the type and concentration of contaminants and detailed site mapping must classify areas of both high and low risk. 3D-ICAS is being developed as a remote system to perform rapid *in situ* analysis of hazardous organic and radionuclide contamination on structural materials. Surface characterization includes identification of potentially dangerous inorganic materials such as asbestos and transite. Real-time remotely operable characterization instrumentation will significantly advance analysis capabilities beyond those currently employed. Chemical analysis plays a vital role throughout the process of decontamination by providing a means to measure progress and to adjust clean-up strategy. Once the clean-up process has been completed the results of chemical analysis will verify that the site is in compliance with federal and local regulations.

The 3D-ICAS configuration consists of a mobile sensor and mapper platform that operates in contaminated areas and an integrated workstation that remains in a safe location. During characterization operations, the mapper, using its coherent laser radar (CLR), determines precise locations used to position the multi-sensor probe located on a robotic arm extending from the platform. The CLR generates 3D facility maps to establish a three-dimensional "world view" within which the robotic sensor system can operate. The operator at the integrated workstation, using displayed 3D map information, plans and directs the selection of surface areas to be characterized and the number of samples for a given area. The system then automatically samples these areas, archives the data (3D location, time, and concentrations of each contaminant), and provides map displays at the workstation showing results of the analysis. The data archive allows regulatory review of the characterization process and assures data integrity. The surface sampling allows the establishment of the bounds of regions exceeding regulatory limits.

In approximately one minute, the high-speed gas chromatograph-mass spectrometer (GC/MS) sensor provides organic analysis, accomplishing detection and fine grain analysis of regulatory concentrations (EPA 1987 spill cleanup policy: $1\mu g/10cm^2$ for high use interior building surfaces). This compares with traditional GC/MS laboratory analysis methods that involve time-consuming sample preparation with associated waste generation. The GC/MS sensor extracts volatile organics directly from contaminated surfaces without contacting the surface. The GC then uses multi-stage focusing to achieve high-resolution insertion into a high-speed gas chromatograph. Detection and additional discrimination are provided by a final stage time-of-flight mass spectrometer (TOF-MS). This high-speed process replaces sample collection and transport as well as hours of solution preparation before injection into an ordinary GC/MS, which typically has a 45-minute run time.

The radionuclide (RN) sensor combines α, β, and γ counting with energy discrimination on the α channel. This sensor combination identifies and quantifies isotopes of specific DOE interest (uranium, plutonium, thorium, technetium, and americium) to regulatory levels in approximately one minute.

The Molecular Vibrational Spectrometry (MVS) sensor characterizes substrate material such as concrete, transite, wood or asbestos. The surface composition information provided by the MVS can be used to provide estimates of the depth of contamination and to optimize the analysis performance of the other contamination detection sensors. The materials composition information augments the surface geometry maps provided by the coherent laser radar, providing a complete 3D worldview for planning and executing robotic D&D operations.

The Integrated Workstation (IWOS) provides data archiving and display and operator interface.

A single mobility platform provides mobility for the robot arm carrying the sensors and for the CLR mapper/tracker. The units may be placed on separate platforms for extended functionality.

Immediately after each one to two minute sample period, the 3D-ICAS sensor output and contamination analysis along with CLR position information is available for near real-time monitoring. After a surface mapping operation is completed, 3D-ICAS will provide 3D displays showing contours of detected contaminant concentrations. 3D-ICAS will further provide permanent measurement data and contaminant level archiving, which assures data integrity and allows straightforward regulatory review of the characterization process before and after D&D operations.

II. SUBSYSTEM DESCRIPTIONS

3D-ICAS is being developed in a three-phase program. Phase I demonstrated the GC/MS sensor system breadboard. Phase II provided integration of the GC and MS subsystems, integration of the multi-sensor probe with the robot arm and the coherent laser radar tracker. The Phase II 3D-ICAS demonstration, involving contamination surface mapping with a CLR guided, robotically maneuvered multi-sensor probe, occurred in November 1995. The Phase III system was integrated and successfully demonstrated on an overhead crane transporter in the fall of 1997 at ORNL. Enhancements and system upgrade will be demonstrated in early 2000.

A. High Speed Gas Chromatograph/Mass Spectrometer Subsystem

Definitive quantitative measurement of organic contamination on building surfaces in less than one minute was the development effort goal.

1. Mass Spectrometer.
Effective High Speed GC/MS performance requires a detector system capable of averaging 100 mass scans over the range of 40-800 amu in 15 milliseconds or less. The time-of-flight (TOF) system is the only full spectrum mass analyzer capable of such a scan rate. The full mass spectrum is necessary for identification of the unknown components of a sample through library searching. Finally, existing TOF technology will allow high repetition scan rates for implementing high duty cycle, high speed operation of the TOF-MS detector when coupled to the High Speed GC. The TOF-MS generates a complete mass spectrum across the mass range 35 to 600 amu every 100 microseconds. An integrated mass spectrum corresponding to 12.8

milliseconds of GC elution time is obtained by averaging 128 mass scans. This integration process yields a signal-to-noise improvement of approximately 11. The ion intensities of the integrated mass spectrum are then summed to generate the total ion chromatogram.

One of the most critical parts of a GC/MS is the interface between the systems. The interface plays the important role both in accommodating the pressure drop from the GC column to the MS ionization source and in enriching the concentration of the analyte in the carrier gas after passing through the interface into the mass spectrometer. In general, removing as much of the carrier gas as possible accommodates the pressure drop. A variety of technologies are available for the removing carrier gas in a GC/MS interface. Briefly, the requirements for a GC/MS interface are high transfer efficiency, no impact on the GC separation, no degradation of compounds, and no preferential removal of compounds or chemical functional groups. In addition to these requirements, the interface in this effort must operate on a fast time scale.

Of all the techniques available, the concept of a molecular beam interface was chosen as optimal. A molecular beam GC to MS interface consists of a heated transfer line to efficiently carry the GC column effluent into a vacuum expansion chamber. The GC column is held in alignment with a skimmer nozzle. The GC column and skimmer are held in close proximity. As the GC column effluent exits the column the lighter carrier gas diffuses away from the sample analyte in the vacuum region. The heavier sample analyte are transferred through the skimmer nozzle in a directed beam with the momentum of the carrier gas. In this approach the carrier gas is removed and the analytes are enriched into a directed beam of molecules. The directed beam of molecules is aligned through the skimmer to intersect the electron gun beam in the ionization source. The introduction of the molecules into the ionization source of the MS in this manner removes the carrier gas, enriches the analyte and generates a directed beam of molecules into the electron gun beam for ionization.

2. GC/MS Processing.

The data from the GC/MS sensor is processed into mass spectra and a total ion chromatogram. The GC/MS data are processed through a National Institutes of Standards and Technology (NIST) Mass Spectral Database, response factor calibration and sample classification algorithms for further data reduction. The electronics technology for the HSGC/MS data acquisition and processing are a high-speed transient digitizer, a high-speed data transfer bus at 20 Mb/sec and a high-speed digital signal processor (DSP). Since the TOF-MS events occur on a nanosecond time scale, the transient digitizer is used to accurately determine the data burst. A mass spectral scale from 35 to 600 amu requires approximately 40 microseconds of data with the individual MS peaks being approximately 15 to 30 nanoseconds wide at the half height point. Thus, to achieve mass spectral resolution of 1 amu at 600 amu, 5 to 10 nanosecond time discrimination is required. The transient digitizer is capable of summing the data burst waveforms in onboard memory to allow signal averaging. At

this point in the data flow, the full mass spectrum is represented as time data with a resolution of 5 nanoseconds. The data records are transferred from the transient digitizer to the digital signal processor over a high-speed data transfer bus at 20 Mb/second. This high-speed transfer is used to recycle the transient digitizer for the next set of data. The high-speed digital signal processor performs many crucial functions in the initial stages of the data processing. These function include the conversion of the time-of-flight data into mass spectral scale data, detection of the occurrence of GC peaks and determination whether the detected GC peak is comprised of one or two compounds, e.g., co-eluting compounds. Once the GC and MS information are extracted from the raw data on the high-speed digital signal processor, the data are transferred to the overall system controller.
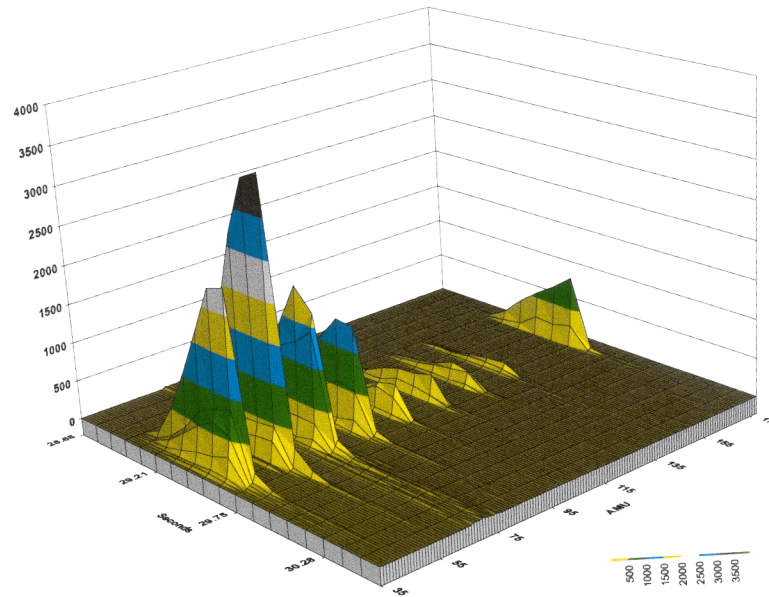

Figure 1 - 3D GC/MS of Hydrocarbon Analyte

B. Radionuclide Sensors

Because the majority of the isotopes are alpha emitters (Am, Th, Pu, U) and one is a beta emitter (Tc), a set of two detectors was determined to best meet the functional needs for the radioisotopes of interest. The radionuclide sensor system includes both a diffused junction silicon detector for alpha emitting isotopes and a sealed gas proportional detector for beta/gamma emitting isotopes. A multichannel analyzer is integrated with the diffused junction detector for discrimination of the alpha isotopes. This design ensures that all the isotopes of interest will be identified. The specific alpha emitting isotopes U^{238}, U^{235}, Pu^{239}, Pu^{242}, Am^{241} and Th^{230} are identified and quantified. The total beta/gamma activity will be reported as Tc^{99}. Isotopic discrimination of beta emitters is not integrated in the current design because the current state of the art instrumentation for beta discrimination would make the MSP weight and size unmanageable. Gamma activity can be discriminated from the beta activity simply by mechanical shuttling of metal plate discriminators.

1. **Alpha Detection**

   The starting point for the alpha detection was Eberline's existing commercial product, the Alpha-6. This device is typically used in air monitoring application for discrimination of low levels of airborne alpha isotopes. The alpha isotopes are discriminated on the basis of pulse height determined from the penetration depth of the particle through the silicon substrate. The data are recorded as a function of energy in the multichannel analyzer. The discrimination is based upon "regions of interest" which are collection of channels summed together to give a response for a particular isotope. A one-inch detector was integrated into the MSP. The associated electronics were integrated into the control software for the sensors.

2. **Beta/Gamma Detection**

   The starting point for the beta/gamma technology was Eberline's Gas Proportional detector and modular detector board used in various commercial products. The gas (argon-carbon dioxide mixture) undergoes ionization by incident radiation. Charge is collected on the anode that is capacitively coupled to a comparator. Thresholds in the comparator discriminate noise and high-energy alpha emission from the beta/gamma response. A two-inch diameter gas proportional tube was integrated into the MSP. A film winder with various thickness of shielding discriminates the gamma radiation from the beta. The associated electronics were integrated into the control software for the sensors.

C. **The Molecular Vibrational Sensor (MVS)**

   The sensor for identification of base materials of interest to DOE (concrete, transite, asbestos, wood, and other organics) is a combination of an infrared (IR) reflection sensor and a Raman scattering sensor. The results obtained with the silica fiber-optic probe analysis of the materials of DOE interest indicated that near infrared (NIR) diffuse reflectance (DR) spectrometry with a silica fiber-optic probe could be used to analyze all materials except those that totally absorb NIR radiation. It was also shown that NIR DR spectrometry could be used to distinguish between the five major types of asbestos. In Phase II it was shown that NIR detectors operating at room temperature did not allow for the spectral features that are important for distinguishing asbestos from analogous samples such as bricks and concrete to be extracted from the baseline noise. Recent advances in room temperature Indium Gallium Arsinide (InGaAs) detectors have allowed University of Idaho (UI) to use NIR DR to identify all the base materials with high reproducibility without the need to resort to the more expensive Extended Near Infrared (ENIR) approach. The use of NIR DR also allows much longer sensor to spectrometer distance and reduced MSP weight.

1. **Near Infrared Spectrometry**

A silica 7-around-1 fiber-optic probe for DR measurements coupled with a room temperature InGaAs detector has been interfaced to an ATI/Mattson Genesis Fourier Transform (FT) spectrometer. The system will cover the range 8000 - 4500 cm^{-1}. The use of FT-NIR is necessitated since several materials have very sharp bands (full width at half height of 8 cm^{-1}) in the NIR (especially asbestos). If classical low resolution dispersive NIR is used, these features are not observed and identification rates for asbestos are lowered. The combination of this probe and detector with the Mattson spectrometer enable high quality spectra to be measured when the probe head was in direct contact with the sample. Typical measurement time is 45 seconds.

2. Raman Spectrometry

Most CCD-Raman spectra were acquired on a Renishaw Raman spectrometer utilizing a microscope as the sampling device in Phase II. The 785-nm diode laser provided a maximum power of 2 mW at the sample. Excitation with gas lasers emitting at 632.8 and 514.5 nm was also tested but fluorescence overwhelmed the Raman spectrum features for most materials of interest. Different lenses allowed standoff distances between 1 and 10 mm to be tested. Single and multiple-scan accumulation spectra were collected at approximately 6 cm^{-1} resolution using CCD detector charge integration times from 1 to 120 seconds. Band positions are accurate to within 2 cm^{-1}. The results obtained with this spectrometer without the fiber-optic probe were sufficiently encouraging to investigate other commercially available Raman spectrometers to which fiber-optic probes had been interfaced. Of these instruments, the one which yielded spectra of solid samples with the highest signal to noise ratio (SNR) was the Kaiser Optical Holoprobe Raman spectrometer. The Kaiser Holoprobe is a state-of-the art NIR Raman spectrometer optimized for use with 785 nm excitation. The system comprises a 300 mW external cavity-stabilized diode laser providing a maximum laser power at the sample of greater than 100 mW, a transmission grating, a CCD detector, and a state-of-the-art fiber-optic probe packaged in a smaller volume than the Renishaw spectrometer. This system offered a usable Raman shift wavenumber range of 50 - 3500 cm^{-1} with 785 nm excitation.

3. Tests for the Appropriate Excitation Wavelength

When the 632.8 nm and the 514.5 nm laser lines were used for excitation, the spectra were so severely affected by molecular and/or atomic luminescence that no Raman bands could be observed above the background. Thus the wavelength for the excitation laser for the MVS Raman probe had to be in the near infrared; in practice the only two wavelengths between which a choice had to be made were 1064 and 785 nm. The 785 nm excitation was selected because of the greater strength of the Raman effect and shorter sample time required at that wavelength.

4. Probe-head Arrangements

It has been shown at the UI that the background radiation caused by the Raman spectrum of silica in the input fiber must be filtered out of the Raman signal from the sample can be significantly degraded or completely obscured. Since a number of

materials of interest in this project (most importantly asbestos) possess crucial vibrational information in the region 800-50 cm^{-1}, the chisel-tip probe approach pioneered at Dow Chemicals and the flat-tip approach proved to be unsuitable for the DOE's specific needs. Because both methods fail to meet the sample-dictated criteria necessary for routine analysis they have not been adopted. The device based on the Kaiser Holoprobe Raman spectrometer with an EIC probe is the best available design (given the MSP weight and size constraints) for studies of weakly Raman scattering solid materials, especially when the wavenumber range below 800 cm^{-1} is important (as it is for asbestos).

5. Software for Identification and Classification

Two types of software for classification and identification have been evaluated: spectral searching and self-organized mapping (SOM) neural networks. In spectral searching, the measured spectrum is compared directly to a library of reference spectra; the absolute difference between the spectrum of the unknown and each reference spectrum is calculated at each wavelength and summed over all wavelengths. The reference spectrum yielding the smallest sum is the best match to the spectrum of the unknown. Spectral searching proves to be most useful when the spectrum of the unknown contains several narrow bands that are easily distinguished from the spectral baseline. In this case, an automated baseline correction routine that has been developed at UI can be applied prior to the application of the spectral searching program.

Unfortunately, the spectra of many of the samples encountered in this project are not of the ideal form for spectral searching. For such samples two other techniques were investigated: principal components analysis (PCA), and self-organized mapping (SOM) neural networks. For both techniques, the dimensionality of the input date is reduced from a fairly large number (the number of data points in each spectrum, often several hundred) to a much smaller number (usually two). In each case, a two-dimensional plot is produced in which it is hoped that samples of a given type will form a cluster in a given region of the plot. In PCA, for example, the ordinates of the plot are the scores of the first two principal components (PC1 and PC2). When the ENIR spectra of a number of samples of brick, concrete, asbestos and polymers are subjected to the PCA, quite poor clustering results. With the use of SOM neural networks, on the other hand, most samples of a given type fell within well-defined regions of the map. Similar results were obtained with NIR spectra. Pattern recognition methods have proved to be more robust than either spectral searching or neural network computing and will form the basis of the classification software.

D. X-RAY FLUORESCENCE (XRF)

In an attempt to provide the most useful information, we are currently evaluating the NITON model 701A X-Ray Fluorescence Spectrometer for addition to the system. The device is capable of determining substrate composition even if the surface is

painted. It is also capable of adding information on toxic metal and substrate identification. It will be tested in the robotic system in the fall 1999.

The starting point for the XRF was NITON corporations model 701A handheld XRF unit with surface identification routines installed. The unit was modified to allow the safety shutter to be opened remotely via a cable release system, and to allow remote operation via computer control of the acquisition software. The built-in battery was replaced with a power supply. Safety from the Cd^{109} source is integrated into the unit.

E. SYSTEM INTEGRATION

3D-ICAS system uses its coherent laser radar (CLR) to obtain 3D facility maps, then uses the CLR generated 3D data to guide a robot arm borne multi-sensor probe along surfaces for contamination detection, 3D mapping and archiving.
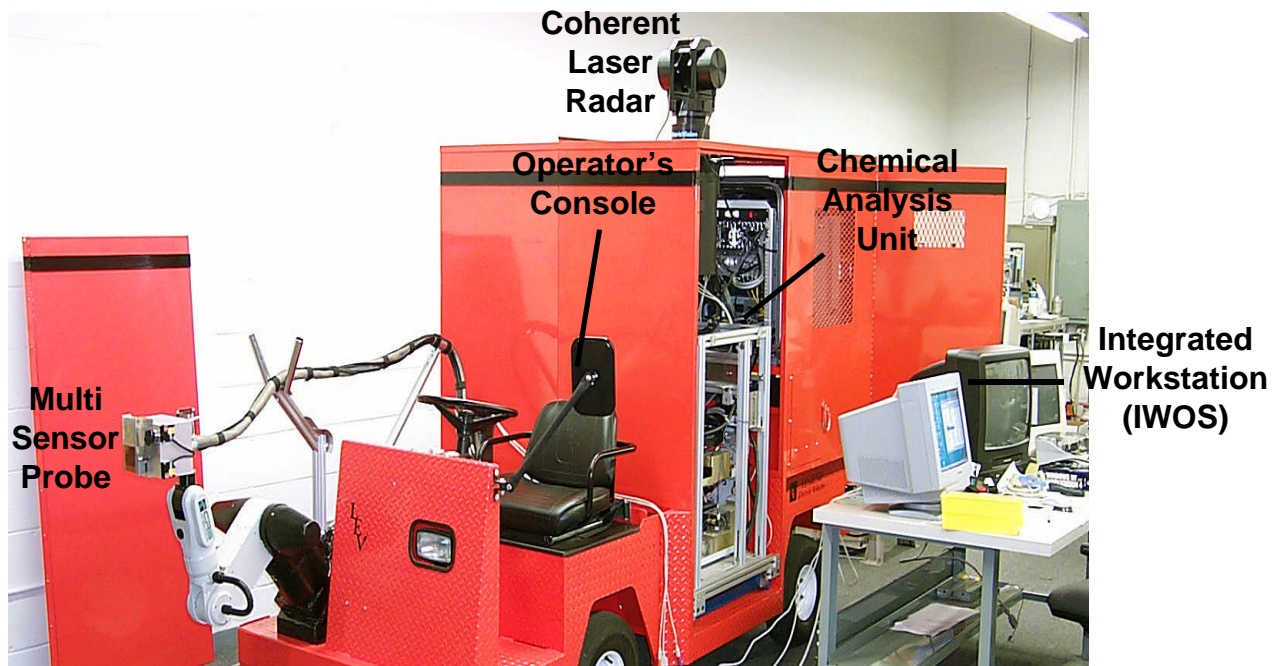

Figure 2 - Prototype Demonstration Unit Under Development

1. Robot Arm

A model 465A robot arm from CRS Robotics (CRS) was selected, programmed and used to manipulate the sensor probe in a limited area. The CRS 465A robot arm has about the same reach as a human's arm. The CRS C500 controller is programmed in the Robot Applied Programming Language (RAPL II), a language developed by CRS, which is a derivative of BASIC and C. In Phase III, the robot arm is being mounted on a mobile platform allowing for full working space access.

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2. Multi-Sensor Probe

The multi-sensor probe comprises the GC sensor head, two MVS probes, two radionuclide sensors, one for alpha particles and one for beta/gamma emissions, and four proximity sensors. The probe is a cube 6 inches on a side and weighs 6 lbs. The system uses commercial proximity sensors mounted at the four corners of the face of the probe. These proximity sensors provide a capability to move the probe up to a flat surface and maintain a standoff distance of 1 mm.

3. Robot Arm Platform

The Phase II system was not required to provide mobility for either the mapper or the robot arm. Mounting each unit on a wheeled tripod and moving them manually provided portability. For the Phase III demonstration, the 3D-ICAS will take advantage of a specially designed mobility platform for both the 3D mapper and the sensor robot arm.

4. Control Strategy

The multi-sensor probe moves to each point specified in the survey list in the order in which they appear in the list generated by the integrated workstation. Movement between points is along a standard sequence of steps. First, the multi-sensor probe moves from the rest position along a straight line to a position near the next survey point, rotating along the way so that the face of the probe is nearly parallel to the surface at the end of the segment. Second, the probe moves toward the surface until the proximity sensors detect the surface. The final approach is accomplished in small steps under control of the proximity sensors. The sensor system is commanded to acquire the contaminant sample and initiate processing. Then the probe moves along a straight line to the arm's rest position. The scan data are processed to provide an accurate estimate of the probe's location and orientation.

5. Database Function

The database capability developed for the 3D-ICAS system serves as a repository for all of the data collected by the system. As the data are gathered, the system automatically archives it for easy retrieval and display by the Integrated Workstation (IWOS). Data are recorded for each survey point. The location of the sensor head is recorded precisely in facility coordinates along with the entire data structure from all of the sensors. The data structure includes raw data from the radionuclide (RN) and High-Speed Gas Chromatograph/Mass Spectrograph (HSGC/MS) sensors, and the Molecular Vibrational Spectrometer (MVS). The data structure also includes processed assignments for four organic compounds provided by the HSGC/MS. Finally, the database holds any specific images generated from the raw 3D mapper data.

6. Data Display: Facility Scene

A facility scene image is generated from the 3D facility frame mapper data by selecting the area to display and an optical viewpoint and converting range from the viewpoint

into a rendered image. The facility scene is rendered in black and white so that subsequent contamination overlays in color will be highlighted.

7. Data Display: Contaminants

When a facility scene is displayed on the IWOS, the database can be queried for related survey information. The data can be overlaid on the facility scene as color-coded points or displayed in one of several different graphical or list windows. Each contaminant is displayed in its own window. The window contains the grayscale facility scene overlaid with contamination levels in color. A box with color codes is placed at each location where a measurement was made. The color of the different sections of the box depends on the concentration of the contaminant. Green, yellow, and red indicate below level of concern, above concern but below need for remediation, and needing remediation for contamination respectively. Default thresholds for defining the three contamination levels are provided; the user may change the thresholds. More detailed displays of contamination data are keyed from the facility scenes with contamination overlays on them. For any contamination display, the operator can select a location to get detailed information about with the mouse. Radionuclide activity levels are displayed in tabular form. Gas chromatograms are displayed in graphical form. Masses of the five organic compound classes are displayed as a bar graph. Secondary displays consisting of mass spectrograms are obtained by clicking on desired peaks in the gas chromatogram; mass spectrograms are displayed as graphs and also in tabular form.
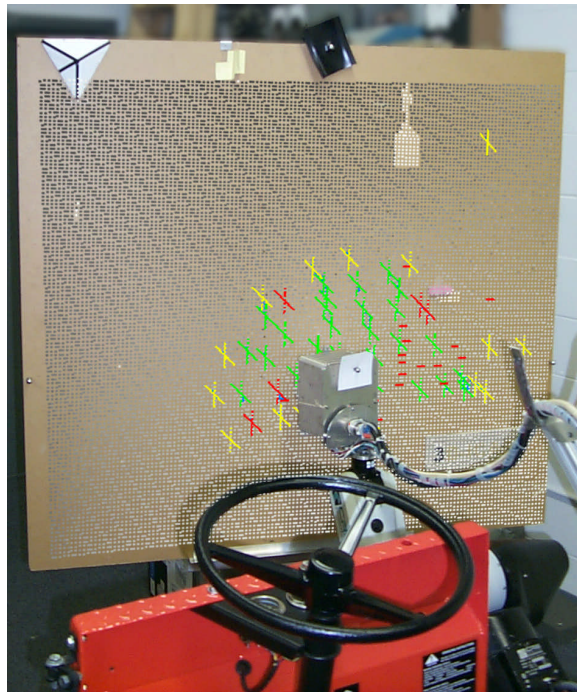

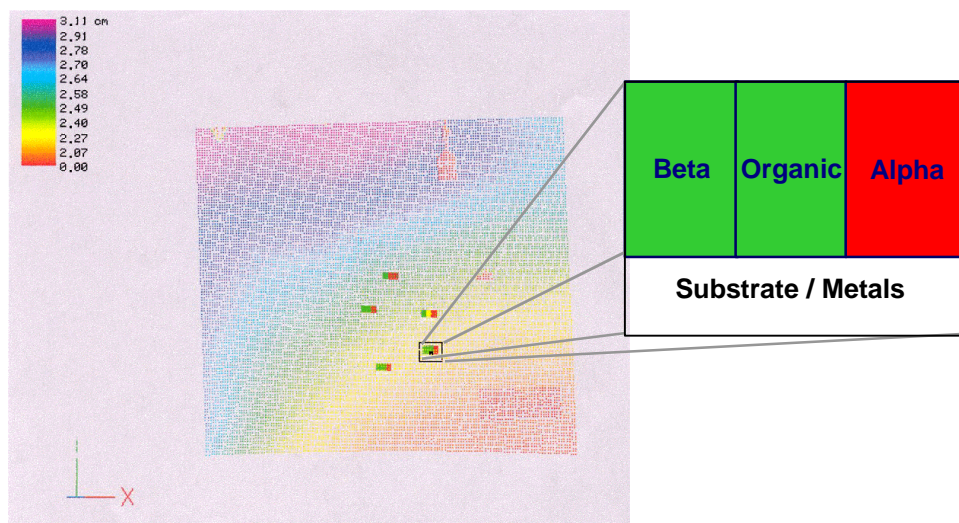**Figure 3 – Overlay of Surveyable Points Map and Site Photograph**

Figure 4 – Top Level Contamination Map

The detailed data package contains the following information:

High Speed GC/MS	Chromatogram to identify organic chemicals
	Mass Spectra, NIST/EPA/NIH Identification Library
	Organic Chemicals classification
Radionuclides	Detects and Quantifies Pu, Am, U, Th, Sr, Tc
X-Ray Fluorescence	Detects Hg, Pb, Cr, U
	Asbestos, Concrete, Metal
Survey Information	Agency, Building, Room, Point of Contact, etc.
	Special Notes

III. CONCLUSIONS

The completed 3D-ICAS will be directly applicable to DOE facility characterization and decontamination and decommissioning. The sensor subsystems of 3D-ICAS represent advances in portable real-time chemical analysis for chemical constituents and dangerous materials of interest to DOE. The CLR 3D mapping subsystem can perform as a critical subsystem over a wide range of DOE robotic applications and industrial metrology applications. The availability of this technology will positively impact DOE clean-up operations in the following ways:

- Improved Performance
- Reduced Cost
- Reduced Health Risk
- Reduction of Environmental Risk

NOMENCLATURE

3D-ICAS	Three Dimensional Characterization and Archiving System
amu	atomic mass units
CCD	Charge coupled detector
CLR	coherent laser radar
CRS	CRS Robotics
D&D	Decontamination and Decommissioning
DOE	Department of Energy
DR	diffuse reflectance
ENIR	extended NIR
FT	Fourier Transform
GC	Gas Chromatograph
GC/MS	gas chromatographs-mass spectrometer
HSGC/MS	High Speed Gas Chromatograph/Mass Spectrometer
IR	Infrared
IWOS	Integrated Workstation
Mb/second	megabits per second
METC	Morgantown Energy Technology Center
MS	Mass spectrometer
MSP	Multi-sensor Probe
MVS	Molecular Vibrational Sensor
NIR	near IR
NIST	National Institutes of Standards and Technology
ORNL	Oak Ridge National Laboratory
PCA	principal components analysis
PCB	polychlorinated biphenyl
RAPL	Robot Applied Programming Language
RN	Radionuclide
SNR	Signal to Noise Ratio
SOM	self-organized mapping
TDI	Thermedics Detection, Inc.
TOF-MS	time-of-flight mass spectrometer
UI	University of Idaho